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(71) Applicant: **DOW GLOBAL TECHNOLOGIES INC.**  
[US/US]; Washington Street, 1790 Building, Midland, MI  
48674 (US).

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(72) Inventor: **MCLEOD, David, G.**; 4631 Deer Springs  
Court, Rochester, MI 48306 (US).

(74) Agent: **DAMOCLES, Nemia, C.**; The Dow Chemical  
Company, Intellectual Property, P.O. Box 1967, Midland,  
MI 48641-1967 (US).

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**WO 03/041985 A1**

(54) Title: **FUEL TANKS AND FUEL TRANSPORT LINES**

(57) Abstract: A fuel tank assembly comprises a fuel tank having a wall with an outer surface and an inner surface, an elongated single or multi-walled tubular body having a first open end and a second open end, the first open end extending outwardly through an opening in the tank wall, and the second open end extending inwardly into the tank and bonded to the tank wall along the periphery of the tank wall opening by an adhesive which bonds to low surface energy plastic materials, the adhesive providing a fuel vapor-tight seal at the interface between the tubular body and tank wall opening, the fuel tank and the tubular body having fuel barrier property.

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## FUEL TANKS AND FUEL TRANSPORT LINES

The present invention relates to plastic fuel tanks and fuel transport lines.

Currently, the attachments of fuel transport  
5 lines to fuel tanks are an area of concern due to cost of the assembly and the excessive emissions of fuel vapor into the atmosphere.

Plastic fuel tanks for automobiles are commonly produced by blow molding process, such as extrusion blow  
10 molding, that is, by extruding a parison into an open mold, closing the mold and blow molding the parison. Extrusion blow molding is a well known process. See, for example, H. G. Fritz "Extrusion Blow Molding," *Plastics Extrusion Technology*, Edited by Friedhelm Hensen, Hanser  
15 Publishers, pp.363-427.

Plastic fuel tanks can also be made by forming or casting a single unit or can be made by joining two or more sections into a finished unit. Further, the tanks can be formed having ports for sensor installation and for  
20 fuel inlet tubes.

Plastic fuel tanks currently use injection molded high density polyethylene (HDPE) nipples that are hot plate welded or spun-welded to the outer tank wall. A single or multi-walled tube is forced-fit over the nipple  
25 and fastened with a mechanical fastener, that is steel strap. The outer end of the tube has, or may have, a quick release fitting that attaches to the steel or polymer tubing that transports the fuel to the engine compartment. The injection molded nipple does not have  
30 fuel barrier property and, therefore, fuel vapors can permeate through the nipples. In addition, fuel vapor can

be lost through the interface between the single or multi-walled tube, the nipple, and the outer wall of the tank.

It would be desirable to provide an assembly of  
35 a fuel tank and fuel transport lines which do not release fuel vapors to the environment.

In a first aspect, the present invention is a fuel tank assembly comprising a fuel tank having a wall with an outer surface and an inner surface, an elongated  
40 single or multi-walled tubular body having a first open end and a second open end, the first open end extending outwardly through an opening in the tank wall, and the second open end extending inwardly into the tank and bonded to the tank wall along the periphery of the tank  
45 wall opening by an adhesive with some barrier properties, the adhesive providing a fuel vapor-tight seal at the interface between the tubular body and tank wall opening, the fuel tank and the tubular body comprising a polymer having fuel barrier property.

50 In a second aspect, the present invention is a fuel tank assembly comprising a fuel tank having a wall with an outer surface and an inner surface, a fuel transport line having a first open end and a second open end, the first open end extending outwardly through an  
55 opening in the tank wall, and the second open end extending inwardly into the tank and bonded to the tank wall along the periphery of the tank wall opening by an adhesive with some barrier properties, the adhesive providing a fuel vapor-tight seal at the interface between  
60 the fuel transport line and tank wall opening, the fuel tank and the fuel transport line comprising a polymer having fuel barrier property.

Fig. 1 is a cross sectional view of a single or multi-walled tubular body which is adapted to be bonded to a conventional plastic fuel tank.

Fig. 2 is a cross sectional view of an alternative embodiment of the single or multi-walled tubular body shown in Fig. 1.

Fig. 3 is a cross sectional view of an assembly comprising the single or multi-walled tubular body shown in Fig. 1 bonded to the inside surface of a fuel tank.

Fig. 4 is a cross sectional view of an assembly comprising a fuel transport line bonded to the inside surface of a fuel tank.

Referring to Figs. 1, 2 and 3, there is shown a single or multi-walled tubular body 10 with a first end 11 and a second end 12. Spaced from second end 12 is a radially and outwardly extending fluted surface 13 and a raised surface 15. Extending from one side of fluted surface 13 to the nearest side of raised surface 15 is bondline 14.

In operation, tubular body 10 is pushed into (Fig. 1) or pulled through the fuel tank (Fig. 2) through a hole cut out of the wall. Fluted surface 13 and raised surface 15 snap fit over tank wall 16. As shown, tubular body 10 is attached to tank wall 16 through fluted surface 13 which is bonded to tank wall 16 along bondline 14 by means of adhesive 17. Raised surface 15 holds tubular body 10 until adhesive 17 is cured to an acceptable green strength. Either bondline 14 or fluted surface 13 is coated with adhesive 17. Adhesive 17 provides a fuel vapor-tight bond between tubular body 10 and tank wall 16. A quick connect may be added to the end of the tubular

body outside the tank. Quick connects are well known in  
95 the art, and are described, for example in U.S. Patent  
5,310,226.

Referring now to Fig. 4, the "tank end" of fuel  
transport lines such as vent lines, fuel line and return  
line, is provided with a radially and outwardly extending  
.00 fluted surface 23 and raised surface 25. Extending from  
one side of fluted surface 23 to the nearest side of  
raised surface 25 is bondline 24.

As used herein, the term "tank end" refers to  
the end of the vent lines, fuel line and return line which  
105 is attached to the fuel tank.

To attach the fuel transport lines to a fuel  
tank, each of the tank end of these lines is press-fit or  
pulled through into drilled or pre-drilled holes in the  
tank until the fluted surface 23 and raised surface 25  
110 snap fit over tank wall 26. Fluted surface 23 is bonded  
to tank wall 26 along bondline 24 by means of adhesive 27.  
The fuel tank is then moved to the next assembly cell or  
to a curing area.

In general, the fuel transport lines are  
115 produced by extrusion, or injection molding, which is  
known in the art. See, for example, U.S. Patents  
6,190,154 and 6,204,312. The fluted end may be added via  
compression molding or through mandrel forming operations  
during a secondary operation.

120 Preferably, the plastic fuel tank, the multi-  
walled tubular body and the fuel transport lines comprise  
a multilayer laminate structure having one or more layers  
of a low energy surface material and one or more layers of  
a polymer having fuel barrier property.

125 More preferably, the plastic fuel tank, the multi-walled tubular body and the fuel transport lines comprise a three-layer laminate structure having two outer layers of a low energy surface material and a core layer of a polymer having fuel barrier property.

30 The low energy surface materials which can be employed in the practice of the present invention include polyolefins such as polyethylene and polypropylene and polytetrafluoroethylene (PTFE).

135 Polyolefins which can be employed in the practice of the present invention for preparing the multilayer laminate structure include polypropylene, polyethylene, and copolymers and blends thereof, as well as ethylene-propylene-diene terpolymers.

140 Preferred polyolefins are polypropylene, linear high density polyethylene (HDPE), heterogeneously-branched linear low density polyethylene (LLDPE) such as DOWLEX™ polyethylene resin (a Trademark of The Dow Chemical Company), heterogeneously branched ultra low linear density polyethylene (ULDPE) such as ATTANE™ ULDPE (a 145 Trademark of The Dow Chemical Company); homogeneously-branched, linear ethylene/ $\alpha$ -olefin copolymers such as TAFMER™ (a Trademark of Mitsui Petrochemicals Company Limited) and EXACT™ (a Trademark of Exxon Chemical Company); homogeneously branched, substantially linear 150 ethylene/ $\alpha$ -olefin polymers such as AFFINITY™ (a Trademark of The Dow Chemical Company) and ENGAGE® (a Trademark DuPont Dow Elastomers L.L.C.) of polyolefin elastomers, which can be prepared as disclosed in U.S. Patents 5,272,236 and 5,278,272; and high pressure, free radical 155 polymerized ethylene polymers and copolymers such as low

density polyethylene (LDPE), ethylene-acrylic acid (EAA) copolymers such as PRIMACOR™ (Trademark of The Dow Chemical Company), and ethylene-vinyl acetate (EVA) copolymers such as ESCORENE™ polymers (a Trademark of  
160 Exxon Chemical Company), and ELVAX™ (a Trademark of E.I. du Pont de Nemours & Co.). The more preferred polyolefins are the homogeneously-branched linear and substantially linear ethylene copolymers with a density (measured in accordance with ASTM D-792) of 0.85 to 0.99 g/cm<sup>3</sup>, a weight  
165 average molecular weight to number average molecular weight ratio (Mw/Mn) from 1.5 to 3.0, a measured melt index (measured in accordance with ASTM D-1238 (190/2.16)) of 0.01 to 100 g/10 min, and an I10/I2 of 6 to 20 (measured in accordance with ASTM D-1238 (190/10)).

170 The most preferred polyolefin is a high density polyethylene. In general, high density polyethylene (HDPE) has a density of at least 0.94 grams per cubic centimeter (g/cc) (ASTM Test Method D-1505). HDPE is commonly produced using techniques similar to the  
175 preparation of linear low density polyethylenes. Such techniques are described in U.S. Patents 2,825,721; 2,993,876; 3,250,825 and 4,204,050. The preferred HDPE employed in the practice of the present invention has a density of from 0.94 to 0.99 g/cc and a melt index of from  
180 0.01 to 35 grams per 10 minutes as determined by ASTM Test Method D-1238.

Polymers having fuel barrier property which can be employed in the practice of the present invention for preparing the plastic fuel tank and the multi-walled  
185 tubular body include polyamides, polytetrafluoroethylene (PTFE), polyamides, fluoroelastomers, polyacetal homopolymers and copolymers, sulfonated and fluorinated

HDPE, ethylene vinyl alcohol polymers and copolymers, hydroxy-functionalized polyethers and polyesters, and  
190 branched polyesters.

Specific examples of polyamides include nylon 6, nylon 66, nylon 610, nylon 9, nylon 11, nylon 12, nylon 6/66, nylon 66/610, and nylon 6/11.

The single-wall tubular body which is bonded to  
195 the tank wall comprises a plastic material, such as polyethylene (also multi wall HDPE extrusions with EvoH barrier), nylon, polyester, or fluoroelastomers, or a metal material, such as steel and aluminum.

The tie layer, also commonly referred to as an  
200 adhesive layer, which can be employed in the practice of the present invention for preparing the multilayer structure is made of an adhesive material, such as a modified polyethylene elastomer. Preferably, the adhesive material is a maleic anhydride grafted polyethylene or  
205 polypropylene such as ADMER™ (Trademark of Mitsui Petrochemicals) adhesive resin or ethylene-vinyl acetate copolymer resins such as ELVAX™ (Trademark of DuPont).

The adhesives which can be employed in the practice of the present invention for attaching the fuel  
210 transport lines to the fuel tank include those adhesives which bond to low energy surface plastic materials, such as the adhesive commercially known as LEA and described in an advertisement in the SPE Plastics Engineering magazine, March 2001 page 22; and adhesives comprising an  
215 amine/organoborane complex, such as those described in a series of patents issued to Skoultchi (U.S. Patent Nos. 5,106,928, 5,143,884, 5,286,821, 5,310,835 and 5,376,746). These patents disclose a two-part initiator system that is



reportedly useful in acrylic adhesive compositions. The  
220 first part of the two-part system includes a stable  
organoborane/amine complex and the second part includes a  
destabilizer or activator such as an organic acid or an  
aldehyde. The organoborane compound of the complex has  
three ligands which can be selected from C<sub>1-10</sub> alkyl groups  
25 or phenyl groups. Useful amines disclosed include  
octylamine, 1,6-diaminohexane, diethylamine, dibutylamine,  
diethylenetriamine, dipropylenediamine, 1,3-  
propylenediamine, and 1,2-propylenediamine.

Other adhesives which can be employed in the  
230 practice of the present invention for attaching plastic  
components to fuel tanks include those adhesives disclosed  
by Zharov et al. in a series of U.S. Patents (U.S.  
5,539,070; U.S. 5,690,780; and U.S. 5,691,065). These  
patents describe polymerizable acrylic compositions which  
235 are particularly useful as adhesives wherein  
organoborane/amine complexes are used to initiate cure.  
The organoboranes used have three ligands attached to the  
borane atom which are selected from C<sub>1-10</sub> alkyl groups and  
phenyl. The amine is an alkanol amine or a diamine where  
240 the first amine group can be a primary or secondary amine  
and the second amine is a primary amine. It is disclosed  
that these complexes are good for initiating  
polymerization of an adhesive which bonds to low surface  
energy substrates.

245 Pocius in a series of patents (U.S. 5,616,796;  
U.S. 5,621,43; U.S. 5,681,910; U.S. 5,686,544; U.S.  
5,718,977; and U.S. 5,795,657) discloses  
amine/organoborane complexes with a variety of amines such  
as polyoxyalkylene polyamines and polyamines which are the

250 reaction product of diprimary amines and compound having  
at least two groups which react with a primary amine.

The most preferred adhesive which can be  
employed in the practice of the present invention for  
attaching the fuel transport lines to the fuel tank is a  
255 class of preferred amines described in copending  
application U.S. Serial No. 09/466321, filed December 17,  
1999. These preferred amines comprise an  
amine/organoborane complex wherein the organoborane is a  
trialkyl borane or alkyl cycloalkyl borane and the amine  
260 is selected from the group consisting of (1) amines having  
an amidine structural component; (2) aliphatic  
heterocycles having at least one nitrogen in the  
heterocyclic ring, wherein the heterocycles may also  
contain one or more nitrogen atoms, oxygen atoms, sulfur  
265 atoms, or double bonds; (3) primary amines which, in  
addition, have one or more hydrogen bond accepting groups  
wherein there are at least two carbon atoms between the  
primary amine and the hydrogen bond accepting group, such  
that due to inter- or intramolecular interactions within  
270 the complex, the strength of the B-N bond is increased;  
and (4) conjugated imines.

Preferably, the trialkyl borane or alkyl  
cycloalkyl borane corresponds to Formula 1:

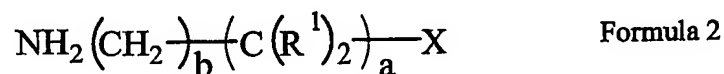


275 wherein B represents Boron; and  $R^2$  is separately in each  
occurrence a  $C_{1-10}$  alkyl,  $C_{3-10}$  cycloalkyl, or two or more of  
 $R^2$  may combine to form a cycloaliphatic ring. Preferably

$R^2$  is  $C_{1-4}$  alkyl, even more preferably  $C_{2-4}$  alkyl, and most preferably  $C_{3-4}$  alkyl.

280 The amine comprises a compound having a primary amine and one or more hydrogen bond accepting groups, wherein there are at least two carbon atoms, preferably at least three, between the primary amine and hydrogen bond accepting groups. Hydrogen bond accepting group means  
 285 herein a functional group that through either inter- or intramolecular interaction with a hydrogen of the borane-complexing amine increases the electron density of the nitrogen of the amine group complexing with the borane. Preferred hydrogen bond accepting groups include primary  
 290 amines, secondary amines, tertiary amines, ethers, halogen, polyethers, and polyamines.

Preferably, the amine corresponds to Formula 2:



wherein:

295  $R^1$  is separately in each occurrence hydrogen or a  $C_{1-10}$  alkyl or  $C_{3-10}$  cycloalkyl;

$X$  is hydrogen bond accepting moiety;  $a$  is an integer of 1 to 10; and  $b$  is separately in each occurrence an integer of 0 to 1, and the sum of  $a$  and  $b$  is from 2 to  
 300 10. Preferably  $R^1$  is hydrogen or methyl. Preferably  $X$  is separately in each occurrence a hydrogen accepting moiety with the proviso that when the hydrogen accepting moiety is an amine it is a tertiary or a secondary amine. More preferably  $X$  is separately in each occurrence  $-N(R^8)_e$ ,

305  $-OR^{10}$ , or a halogen wherein  $R^8$  is separately in each occurrence  $C_{1-10}$  alkyl,  $C_{3-10}$  cycloalkyl or  $-(C(R^1)_2)_d-W$ ;  $R^{10}$  is separately in each occurrence,  $C_{1-10}$  alkyl,  $C_{3-10}$  cycloalkyl, or  $-(C(R^1)_2)_d-W$ ; and  $e$  is 0, 1, or 2. More preferably  $X$  is  $-N(R^8)_2$  or  $-OR^{10}$ . Preferably,  $R^8$  and  $R^{10}$  are

310  $C_{1-4}$  alkyl or  $-(C(R^1)_2)_d-W$ , more preferably  $C_{1-4}$  alkyl and most preferably methyl.  $W$  is separately in each occurrence hydrogen or  $C_{1-10}$  alkyl or  $X$  and more preferably hydrogen or  $C_{1-4}$  alkyl. Preferably,  $a$  is 1 or greater and more preferably 2 or greater. Preferably  $a$  is 6 or less,

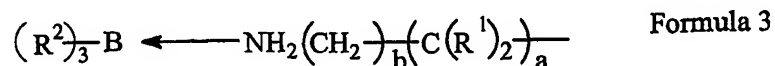
315 and most preferably 4 or less. Preferably,  $b$  is 1. Preferably, the sum of  $a$  and  $b$  is an integer 2 or greater and most preferably 3 or greater. Preferably the sum of  $a$  and  $b$  are 6 or less and more preferably 4 or less. Preferably  $d$  is separately in each occurrence an integer

320 of 1 to 4, more preferably 2 to 4, and most preferably 2 to 3. Among preferred amines corresponding to Formula 2 are dimethylaminopropyl amine, methoxypropyl amine, dimethylaminoethylamine, dimethylaminobutylamine, methoxybutyl amine, methoxyethyl amine, ethoxypropylamine, propoxypropylamine, amine terminated polyalkylene ethers

325 (such as trimethylolpropane tris(poly(propyleneglycol), amine-terminated) ether), aminopropylmorpholine, isophoronediamine, and aminopropylpropanediamine.

In one embodiment the preferred amine complex

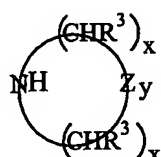
330 corresponds to Formula 3:



wherein  $R^1$ ,  $R^2$ ,  $X$ ,  $a$  and  $b$  are as defined hereinbefore.

In another embodiment the amine is an aliphatic heterocycle having at least one nitrogen in the  
 335 heterocycle. The heterocyclic compound may also contain one or more of nitrogen, oxygen, sulfur or double bonds.

In addition, the heterocycle may comprise multiple rings wherein at least one of the rings has a nitrogen in the ring. Preferably the aliphatic  
 340 heterocyclic amine corresponds to Formula 4:



Formula 4

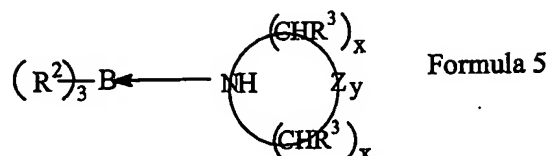
wherein:

$R^3$  is separately in each occurrence hydrogen, a  $C_{1-10}$  alkyl or  $C_{3-10}$  cycloalkyl;

345  $Z$  is separately in each occurrence oxygen or  $NR^4$  wherein  $R^4$  is hydrogen,  $C_{1-10}$  alkyl, or  $C_{6-10}$  aryl or alkaryl;

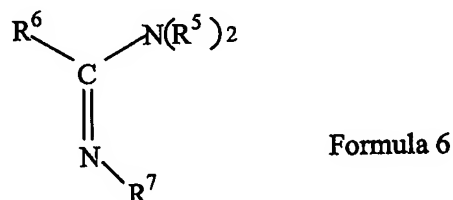
$x$  is separately in each occurrence an integer of 1 to 10, with the proviso that the total of all  
 350 occurrences of  $x$  should be from 2 to 10; and  $y$  is separately in each occurrence 0 or 1. Preferably,  $R^3$  is separately in each occurrence hydrogen or methyl. Preferably  $Z$  is  $NR^4$ . Preferably,  $R^4$  is hydrogen or  $C_{1-4}$  alkyl, and more preferably hydrogen or methyl. Preferably  
 355  $x$  is from 1 to 5 and the total of all the occurrences of  $x$  is 3 to 5. Preferred compounds corresponding to Formula 4 include morpholine, piperidine, pyrrolidine, piperazine, 1,3,3-trimethyl 6-azabicyclo[3.2.1] octane, thiazolidine, homopiperazine, aziridine, 1,4-diazabicyclo[2.2.2]octane  
 360 (DABCO), 1-amino-4-methylpiperazine, and 3-pyrroline.

Complexes using aliphatic heterocyclic amines preferably correspond to Formula 5:



wherein  $R^2$ ,  $R^3$ ,  $Z$ ,  $x$  and  $y$  are as defined hereinbefore.

365 In yet another embodiment, the amine which is complexed with the organoborane is an amidine. Any compound with amidine structure wherein the amidine has sufficient binding energy as described hereinbefore with the organoborane, may be used. Preferable amidine  
370 compounds correspond to Formula 6:

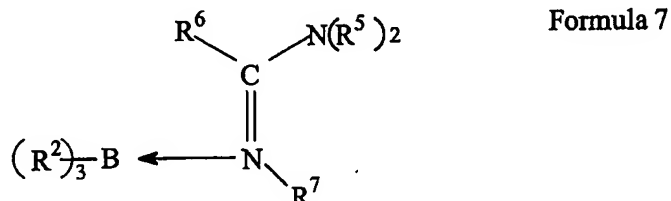


wherein:

$R^5$ ,  $R^6$ , and  $R^7$  are separately in each occurrence  
375 hydrogen, a  $C_{1-10}$  alkyl or  $C_{3-10}$  cycloalkyl; two or more of  $R^5$ ,  $R^6$ , and  $R^7$  may combine in any combination to form a ring structure, which may have one or more rings. Preferably  $R^5$ ,  $R^6$  and  $R^7$  are separately in each occurrence hydrogen,  $C_{1-4}$  alkyl or  $C_{5-6}$  cycloalkyl. Most preferably  $R^7$  is H or  
380 methyl. In the embodiment where two or more of  $R^5$ ,  $R^6$  and  $R^7$  combine to form a ring structure the ring structure is preferably a single or a double ring structure. Among

preferred amidines are 1,8-diazabicyclo[5,4]undec-7-ene;  
 tetrahydropyrimidine; 2-methyl-2-imidazoline; and  
 385 1,1,3,3-tetramethylguanidine.

The organoborane amidine complexes preferably  
 correspond to Formula 7:



wherein R<sup>2</sup>, R<sup>5</sup>, R<sup>6</sup> and R<sup>7</sup> are as defined earlier.

390 In yet another embodiment, the amine which is  
 complexed with the organoborane is a conjugated imine.  
 Any compound with a conjugated imine structure, wherein  
 the imine has sufficient binding energy as described  
 hereinbefore with the organoborane, may be used. The  
 395 conjugated imine can be a straight- or branched-chain  
 imine or a cyclic imine. Preferable imine compounds  
 correspond to Formula 8:

Formula 8



400 wherein Y is independently in each occurrence hydrogen,  
 N(R<sup>4</sup>)<sub>2</sub>, OR<sup>4</sup>, C(O)OR<sup>4</sup>, halogen or an alkylene group which  
 forms a cyclic ring with an R<sup>7</sup> or R<sup>9</sup>. R<sup>4</sup> is hydrogen, C<sub>1-10</sub>  
 alkyl, or C<sub>6-10</sub> aryl or alkaryl. Preferably R<sup>4</sup> is hydrogen  
 or methyl. R<sup>7</sup> is as described previously. R<sup>9</sup> is  
 405 independently in each occurrence hydrogen, Y, C<sub>1-10</sub> alkyl,

C<sub>3-10</sub> cycloalkyl-, (C(R<sup>9</sup>)<sub>2</sub>-(CR<sup>9</sup>=CR<sup>9</sup>)<sub>c</sub>-Y or two or more of R<sup>9</sup> can combine to form a ring structure provided the ring structure is conjugated with respect to the double bond of the imine nitrogen; and c is an integer of from 1 to 10.

410 Preferably, R<sup>9</sup> is hydrogen or methyl.

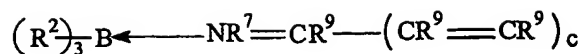
Y is preferably N(R<sup>4</sup>)<sub>2</sub>, or OR<sup>4</sup>, or an alkylene group which forms a cyclic ring with R<sup>7</sup> or R<sup>9</sup>. Y is more preferably N(R<sup>4</sup>)<sub>2</sub> or an alkylene group which forms a cyclic ring with R<sup>7</sup> or R<sup>9</sup>. Preferably, c is an integer of from 1  
415 to 5, and most preferably 1. Among preferred conjugated imines useful in this invention are 4-dimethylaminopyridine; 2,3-bis(dimethylamino)-cyclopropeneimine; 3-(dimethylamine)acroleinimine; 3-(dimethylamino)methacroleinimine.

420 Among preferred cyclic imines are those corresponding to the following structures

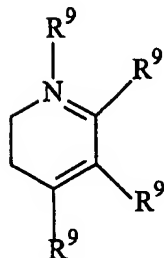


The complexes with the conjugated imines  
425 preferably correspond to Formula 9:





Formula 9



wherein  $R^2$ ,  $R^7$ ,  $R^9$ ,  $c$  and  $Y$  are as defined hereinbefore.

The molar ratio of amine compound to borane  
 430 compound in the complex is relatively important. In some  
 complexes if the molar ratio of amine compound to  
 organoborane compound is too low, the complex is  
 pyrophoric. Preferably the molar ratio of amine compound  
 to organoborane compound is from 1.0:1.0 to 3.0:1.0.  
 435 Below the ratio of 1.0:1.0 there may be problems with  
 polymerization, stability of the complex and for adhesive  
 uses, adhesion. Greater than a 3.0:1.0 ratio may be used  
 although there is no benefit from using a ratio greater  
 than 3.0:1.0. If too much amine is present, this may  
 440 negatively impact the stability of the adhesive or polymer  
 compositions. Preferably the molar ratio of amine  
 compound to organoborane compound is from 2.0:1.0 to  
 1.0:1.0.

Polymerizable compounds which may be used in the  
 445 polymerization compositions of the adhesive include  
 acrylate and/or methacrylate based compounds, with  
 methylmethacrylate, butylmethacrylate, 2-  
 ethylhexylmethacrylate, isobornylmethacrylate,  
 tetrahydrofurfuryl methacrylate, and  
 450 cyclohexylmethylmethacrylate as the most preferred.

Each of the polymers forming the layers of the multilayer laminate structure of the present invention may contain various additives in an amount that does not adversely affect the desired properties of the polymers.

455 Examples of such additives include antioxidants, ultraviolet light absorbers, thermal processing stabilizers, colorants, lubricants, flame retardants, impact modifiers, plasticizers, antistatic agents, pigments, and nucleating agents and fillers, such as

460 zeolite, talc, and calcium carbonate. The method of incorporating the additives is not critical. The additives can conveniently be added to the polymer prior to preparing the multilayer laminate structure. If the polymer is prepared in solid form, the additives can be

465 added to the melt prior to preparing the multilayer laminate structure.

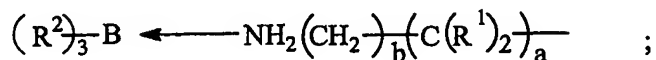
WHAT IS CLAIMED IS:

470 1. A fuel tank assembly comprising a fuel tank  
having a wall with an outer surface and an inner surface,  
an elongated single or multi-walled tubular body having a  
first open end and a second open end, the first open end  
extending outwardly through an opening in the tank wall,  
475 and the second open end extending inwardly into the tank  
and bonded to the tank wall along the periphery of the  
tank wall opening by an adhesive which bonds to low  
surface energy plastic materials, the adhesive providing a  
fuel vapor-tight seal at the interface between the tubular  
480 body and tank wall opening, the fuel tank and the tubular  
body comprising a polymer having fuel barrier property.

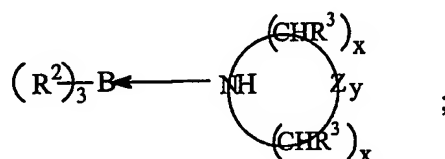
2. The fuel tank assembly of Claim 1 wherein  
the adhesive is an amine/organoborane complex.

3. The fuel tank assembly of Claim 2 wherein the  
485 organoborane compound of the complex is a trialkyl borane  
or alkyl cycloalkyl borane and the amine compound is  
selected from the group consisting of (1) amines having an  
amidine structural component; (2) aliphatic heterocycles  
having at least one nitrogen in the heterocyclic ring,  
490 wherein the heterocyclic compound may also contain one or  
more nitrogen atoms, oxygen atoms, sulfur atoms, or double  
bonds in the heterocycle; (3) primary amines which, in  
addition, have one or more hydrogen bond accepting groups  
wherein there are at least two carbon atoms between the  
495 primary amine and the hydrogen bond accepting group, such  
that due to inter- or intramolecular interactions within  
the complex, the strength of the B-N bond is increased;  
and (4) conjugated imines.

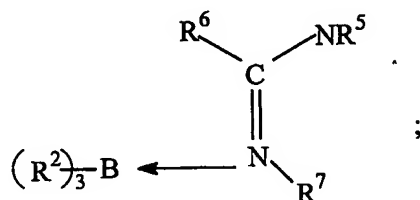
4. The fuel tank assembly of Claim 2 wherein  
 500 the complex of the organoborane and the primary amine  
 corresponds to the formula



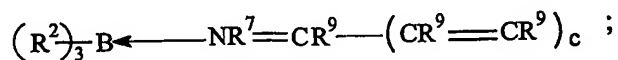
the organoborane heterocyclic amine complex corresponds  
 to the formula



505 the organoborane amidine complex corresponds to the  
 formula



and the organoborane conjugated imine complex  
 corresponds to the formula



510

wherein:

B is boron;  $R^1$  is separately in each occurrence  
 hydrogen, a  $C_{1-10}$  alkyl or  $C_{3-10}$  cycloalkyl;

R<sup>2</sup> is separately in each occurrence a C<sub>1-10</sub> alkyl,  
515 C<sub>3-10</sub> cycloalkyl or two or more of R<sup>2</sup> may combine to form a  
cycloaliphatic ring structure;

R<sup>3</sup> is separately in each occurrence hydrogen, a  
C<sub>1-10</sub> alkyl or C<sub>3-10</sub> cycloalkyl; R<sup>4</sup> is separately in each  
occurrence hydrogen, C<sub>1-10</sub> alkyl, C<sub>3-10</sub> cycloalkyl, C<sub>6-10</sub> aryl  
520 or alkaryl; R<sup>5</sup>, R<sup>6</sup>, and R<sup>7</sup> are separately in each  
occurrence hydrogen, C<sub>1-10</sub> alkyl, C<sub>3-10</sub> cycloalkyl, or two or  
more of R<sup>5</sup>, R<sup>6</sup> and R<sup>7</sup> in any combination can combine to  
form a ring structure which can be a single ring or a  
multiple ring structure and the ring structure can include  
525 one or more of nitrogen, oxygen or unsaturation in the  
ring structure; R<sup>9</sup> is independently in each occurrence  
hydrogen, C<sub>1-10</sub> alkyl or C<sub>3-10</sub> cycloalkyl, Y, -(C(R<sup>9</sup>))<sub>2</sub>-  
(CR<sup>9</sup>=CR<sup>9</sup>)<sub>c</sub>-Y or two or more of R<sup>9</sup> can combine to form a ring  
structure, or one or more of R<sup>9</sup> can form a ring structure  
530 with Y provided the ring structure is conjugated with  
respect to the double bond of the imine nitrogen; X is a  
hydrogen-bond accepting group with the proviso that where  
the hydrogen bond accepting group is an amine it must be  
secondary or tertiary;

535 Y is independently in each occurrence hydrogen,  
N(R<sup>4</sup>)<sub>2</sub>, OR<sup>4</sup>, C(O)OR<sup>4</sup>, a halogen or an alkylene group which  
forms a cyclic ring with R<sup>7</sup> or R<sup>9</sup>; Z is separately in each  
occurrence oxygen or -NR<sup>4</sup>; a is separately in each  
occurrence an integer of from 1 to 10; b is separately in  
540 each occurrence 0 or 1, with the proviso that the sum of a  
and b should be from 2 to 10; c is separately in each  
occurrence an integer of from 1 to 10; x is separately in  
each occurrence an integer of 1 to 10, with the proviso

that the total of all occurrences of x is from 2 to 10;  
545 and y is separately in each occurrence 0 or 1.

5. The fuel tank assembly of Claim 2 wherein the organo borane/amine complex comprises an aliphatic heterocyclic amine which is a five or six-membered heterocyclic compound.

550 6. The fuel tank assembly of Claim 2 wherein the organo borane compound of the complex has three ligands selected from C<sub>1-10</sub> alkyl groups or phenyl groups, and the amine compound is selected from 1,6-diaminohexane, diethylamine, dibutylamine, diethylenetriamine,  
555 dipropylenediamine, 1,3-propylenediamine, and 1,2-propylene-diamine.

7. The fuel tank assembly of Claim 2 wherein the organoborane compound of the complex has three ligands attached to the borane atom and which are selected from C<sub>1-10</sub> alkyl groups and phenyl and the amine compound is an  
560 alkanol amine or a diamine wherein the first amine group is a primary or secondary amine and the second amine is a primary amine.

8. The fuel tank assembly of Claim 2 wherein  
565 the amine compound of the complex is a polyoxyalkylene polyamine or a polyamine which is the reaction product of a diprimary amine and a compound having at least two groups which react with a primary amine.

9. The fuel tank assembly of Claim 1 wherein  
570 the low surface energy plastic material is a polyolefin.

10. The fuel tank assembly of Claim 9 wherein the polyolefin is selected from the group consisting of polyethylene, polypropylene and polytetrafluoroethylene.

11. The fuel tank assembly of Claim 1 wherein  
575 the polymer having fuel barrier property is selected from  
the group consisting of polyamides, fluoroelastomers,  
polyacetal homopolymers and copolymers, sulfonated and  
fluorinated HDPE, ethylene vinyl alcohol polymers and  
copolymers, hydroxy-functionalized polyethers and  
580 polyesters, and branched polyesters.

12. The fuel tank assembly of Claim 1 wherein  
the fuel tank is a three-layer laminate structure  
comprising two outer layers of a low energy surface  
material and a core layer of a polymer having fuel barrier  
585 property.

13. The fuel tank assembly of Claim 12 wherein  
the low energy surface material is polyethylene and the  
polymer having fuel barrier property is selected from the  
group consisting of polyamides, fluoroelastomers,  
590 polyacetal homopolymers and copolymers, sulfonated and  
fluorinated HDPE, ethylene vinyl alcohol polymers and  
copolymers, hydroxy-functionalized polyethers and  
polyesters, and branched polyesters.

14. The fuel tank assembly of Claim 1 wherein  
595 the tubular body has a first end and a second end, a  
radially and outwardly extending fluted surface and a  
raised surface spaced from the second end, and a bondline  
extending from one side of the fluted surface to the  
nearest side of the raised surface.

600 15. A fuel tank assembly comprising a fuel tank  
having a wall with an outer surface and an inner surface,  
a single or multi-walled fuel transport line having a  
first open end and a second open end, the first open end  
extending outwardly through an opening in the tank wall,

605 and the second open end extending inwardly into the tank  
and bonded to the tank wall along the periphery of the  
tank wall opening by an adhesive which bonds to low  
surface energy plastic materials, the adhesive providing a  
fuel vapor-tight seal at the interface between the fuel  
610 transport line and tank wall opening, the fuel tank and  
the fuel transport line comprising a polymer having fuel  
barrier property.

16. The fuel tank assembly of Claim 15 wherein  
the fuel transport line has a first end and a second end,  
615 a radially and outwardly extending fluted surface and a  
raised surface spaced from the second end, and a bondline  
extending from one side of the fluted surface to the  
nearest side of the raised surface.

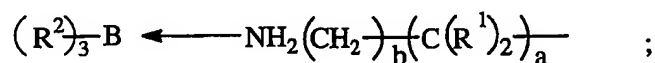


## AMENDED CLAIMS

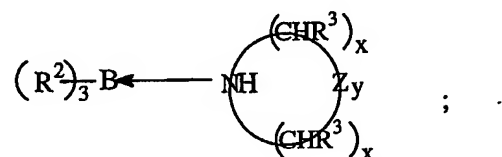
[received by the International Bureau on 17 October 2002 (17.10.02);

CLAIM 4 REPLACED

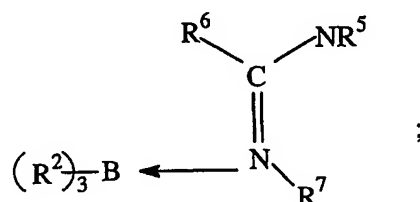
4. The fuel tank assembly of Claim 2 wherein the complex of the organoborane and the primary amine corresponds to the formula



the organoborane heterocyclic amine complex corresponds to the formula

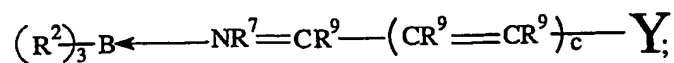


the organoborane amidine complex corresponds to the formula



and the organoborane conjugated imine complex corresponds to the formula

wherein:



B is boron;  $R^1$  is separately in each occurrence hydrogen, a  $C_{1-10}$  alkyl or  $C_{3-10}$  cycloalkyl;

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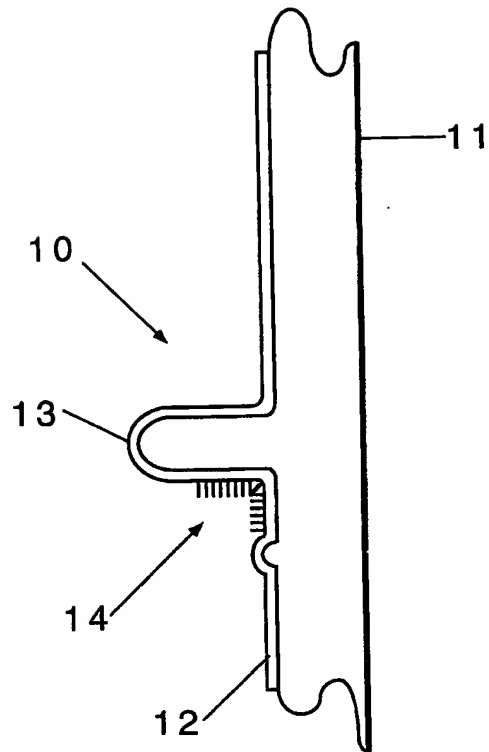


Fig. 1

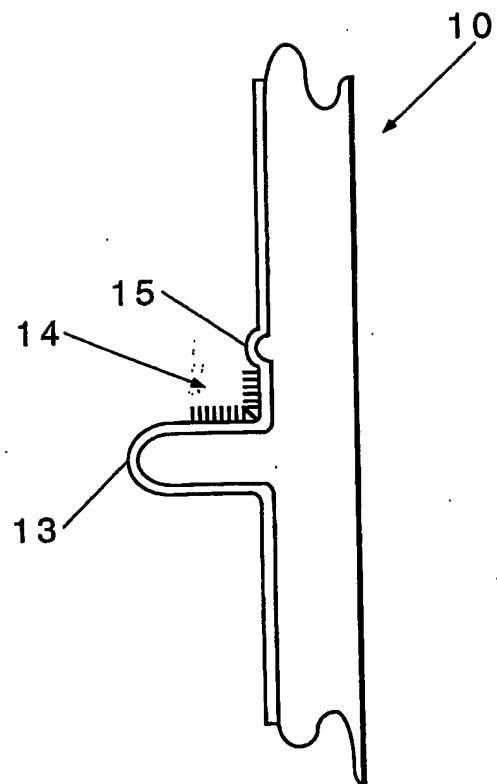
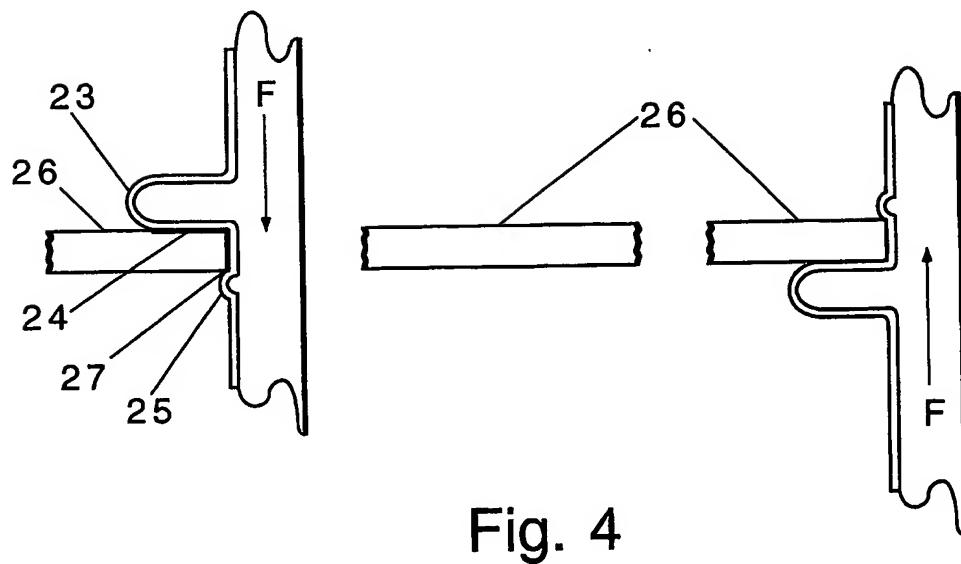
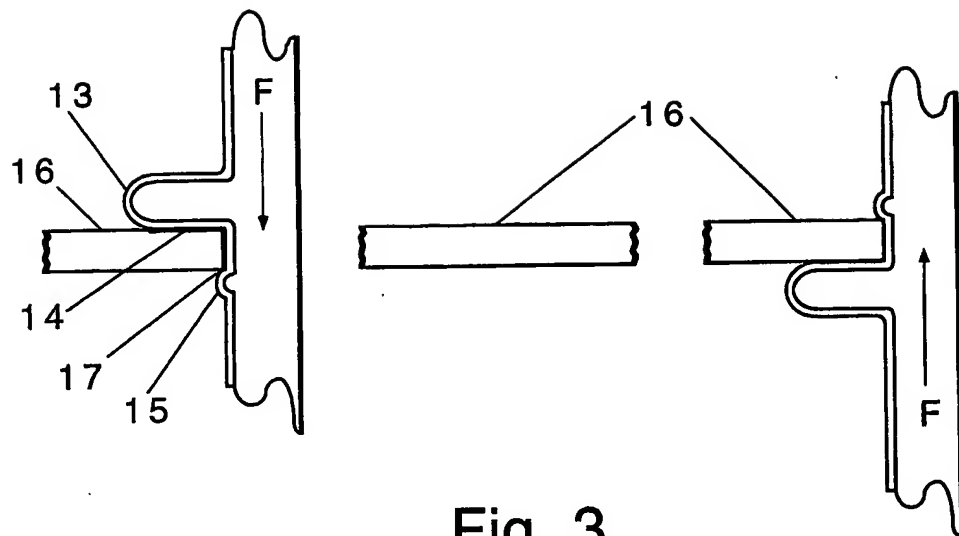


Fig. 2

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# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/US 02/13975

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 7 B60K15/073 B60K15/04

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 B60K F16L C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

PAJ, EPO-Internal, WPI Data

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	PATENT ABSTRACTS OF JAPAN vol. 011, no. 296 (M-627), 25 September 1987 (1987-09-25) & JP 62 089584 A (TOYOTA MOTOR CORP; OTHERS: 02), 24 April 1987 (1987-04-24) abstract	1-16
A	DE 199 07 736 A (VOLKSWAGENWERK AG) 24 August 2000 (2000-08-24) claims 1-3; figures	1-16
A	US 5 690 780 A (KRASNOV DECEASED JURY N ET AL) 25 November 1997 (1997-11-25) cited in the application claim 1	1-16

☐ Further documents are listed in the continuation of box C.

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Date of the actual completion of the international search

30 August 2002

Date of mailing of the international search report

16/09/2002

Name and mailing address of the ISA  
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NL - 2280 HV Rijswijk  
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Authorized officer

Schlicke, B

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 02/13975

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
JP 62089584	A	24-04-1987	NONE	
DE 19907736	A	24-08-2000	DE 19907736 A1	24-08-2000
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			US 5691065 A	25-11-1997
			US 6284381 B1	04-09-2001

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